REPORT DOCUMENTATION PAGE

Form Approved
OMB No 0704-0188

AD-A281 662

n is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, ...ting and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this ...cing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson ...nd to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington DC 20503

2. REPORT DATE
3-1-94

3. REPORT TYPE AND DATES COVERED
Final report 2-22-93 to 2-21-94

4. TITLE AND SUBTITLE
Excimer emission from alkali diatomic and alkaline-earth-noble gas molecules

5. FUNDING NUMBERS
DAAH04-93-G-0063

6. AUTHOR(S)
John Huennekens

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Lehigh University
Dept. of Physics
16 Memorial Dr. East
Bethlehem, PA 18015

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
ARO 30440.4-PH

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

npx 94-21102

13. ABSTRACT (Maximum 200 words)

We have carried out a series of experiments to study collisional and radiative processes in alkali metal and alkaline-earth vapors. Our principal interest has been to understand fundamental atom-atom, atom-molecule, and atom-light interactions in order to model alkali diatomic and alkaline-earth excimer systems for possible laser applications. During this funding period, we have concentrated on collisional excitation transfer, quenching and diffusion processes involving the metastable 6s5d ^3D_J levels of barium. We have also investigated alkali and alkaline-earth vapors as media for observing low threshold nonlinear optical processes.

DTIC QUALITY INSPECTED 5

94 7 11 047

14. SUBJECT TERMS
alkali molecules, barium-noble gas interactions, coherent emission

15. NUMBER OF PAGES
6

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

Final Report - ARO grant DAAH04-93-G-0063

During the one year period funded by this grant, we have made substantial progress on several projects of interest. Since the annual progress report, covering the period 1 January 1993 - 31 December 1993 (i.e., most of the period funded by this grant) was submitted only last month, the present final report is largely redundant with that earlier report.

One area of continuing interest to us is the study of collisional and radiative processes occurring in metastable barium - noble-gas mixtures. The lowest excited states of barium are the 6s5d ^3D_J levels which are metastable since radiation from them to the ground state $6s^2\ ^1S_0$ is dipole forbidden. However, these levels are easily populated through the optical pumping channel $6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1 \rightarrow 6s5d\ ^3D_{1,2}$. We have found that more than 60% of the ground state atoms can be transferred to the metastable levels using either a pulsed or cw pump laser. Much of our early work on this system involved the development of techniques to monitor populations in the various levels in a quantitative fashion. In particular, we carried out detailed collision broadened lineshape measurements (with argon perturbers) for transitions whose lower level is one of the barium metastable ^3D_J states [Phys. Rev. A <u>46</u>, 2642 (1992)]. More recently, similar studies on the $6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1$ intercombination line have been completed [Phys. Rev. A <u>47</u>, 3097 (1993)]. The latter is important since a great deal of controversy exists over the barium vapor pressure curve, and such lineshape studies allow us to determine barium densities using simple optical measurements. With these techniques, we can now monitor populations in any of the low lying states of barium over a wide range of conditions.

When a pulsed laser is used to optically pump the barium atoms to the metastable levels, we can carry out time-resolved studies of a number of interesting processes. In this technique, we still use a single-mode cw laser to monitor the metastable level populations by measuring the absorption of the cw beam when the laser is tuned near one of the transitions for which we have detailed lineshape information. By examining this absorption as a function of time following the firing of the pulsed pump laser, we can map out populations (in absolute units) in a completely time-resolved fashion. During the last year, we have used this technique to study excitation transfer among the metastable levels due to collisions with argon perturbers, and quenching of the metastables by molecular nitrogen. These results are reported in the enclosed manuscript "Excitation transfer among, and quenching of, the barium 6s5d ^3D_J metastable levels due to collisions with argon, nitrogen, and barium perturbers" which has been submitted to Phys. Rev. A. By displacing the cw probe beam parallel to the counterpropagating pump, we can also obtain detailed spatial information. This has led us to study the diffusion of the metastable atoms through argon. A manuscript on this latter work is in preparation. The barium work described so far, including the cw lineshape measurements, the pulsed mixing and quenching studies, and the diffusion measurements form the core of the Ph.D. dissertation of Ed Ehrlacher.

More recently, a new doctoral student, Ms. Raychel Brown, has joined our group. As a first project, she has carried out a study of diffusion of ground state barium atoms through argon gas. The technique here is similar to that used with the metastable states except that the weak cw probe is also tuned to the $6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1$ intercombination

line. Thus the pump laser depletes the ground state, and the probe laser interrogates the spatial hole in the ground state population distribution which fills and widens due to diffusion. In this manner we hope to get an accurate ground state diffusion constant which we can compare to the metastable level diffusion constant. These values are needed for the accurate modeling of laser pumped barium vapors. In particular, light-induced kinetic effects such as light-induced drift depend upon the difference between the ground and excited state diffusion cross sections. We anticipate that Ms. Brown will extend these measurements for both the ground and metastable levels to include the other inert perturber gases. Thus, the most promising candidates for the study of light-induced kinetic effects in barium will become apparent.

In another project, we have become interested in possibilities of observing nonlinear optical processes at unusually low thresholds in barium vapor. Here, we consider both the nonlinear absorption and refractive index variation when a laser tuned near the $6s^2\ {}^1S_0 \rightarrow 6s6p\ {}^3P_1$ transition is incident upon a barium vapor. For a two-level atom, at low incident laser powers, the absorption is linear (i.e., doubling the laser intensity doubles the number of absorbed photons). As the intensity increases, net absorption fails to increase at the same rate due to depletion of the lower state and the competition of stimulated emission from the upper state. This process is called saturation and is characterized by a parameter called the saturation intensity I_{sat}. Incident intensities on the order of I_{sat} produce nonlinear responses which can lead to such interesting phenomena as four-wave mixing, two-beam coupling, and optical bistability. Due to the presence of the low lying metastable $6s5d\ {}^3D_J$ levels, barium acts as a three level system when the laser is tuned near the $6s^2\ {}^1S_0 \rightarrow 6s6p\ {}^3P_1$ transition. Density matrix or rate equations show that the metastable levels act as a trap for ground state population, resulting in a much lower saturation intensity than for a two-level atom. Our goal is to use this system to produce nonlinear optical phenomena (such as degenerate four-wave mixing) at very low incident beam intensities. To date, we have measured saturation intensities in barium, cesium, rubidium, and potassium. The three alkali metals are also of much interest in this context because the ground state hyperfine level splitting in the heavy alkalis (cesium and rubidium) is sufficiently large that these atoms also act as three level systems with low saturation intensities. However in potassium, the hyperfine splitting is less than the Doppler widths of the lines. Therefore potassium must be modeled as a two level system and has a much larger saturation intensity (we have recently confirmed this result experimentally). Modeling of all of these systems, including lineshape and attenuation effects, is currently in progress. Four-wave mixing has been observed in rubidium, and we plan to extend these results to the other cases. We are particularly interested in understanding the role of diffusion and quenching of the metastable atoms in the nonlinear response of barium. In particular, it seems to us that if the trap level atoms effectively decay only by diffusion, then nonlinear processes such as four-wave mixing and two-beam coupling, which depend on the existence of a refractive-index or population "grating", should not be observable. On the other hand, processes such as optical bistability should be observable under the same conditions. By addition of a quenching gas such as nitrogen, we can switch between conditions where the effective decay mechanism is quenching as opposed to diffusion. These studies have been undertaken by an excellent graduate student, Jon Sagle, who will continue with them during the course of

the next year. An outstanding undergraduate, Ms. Amy Perkins from Drew University, also joined in this work last summer as part of our department's NSF sponsored "Research Experiences for Undergraduates" program.

Publications resulting from ARO grant DAAH04-93-G-0063

E. Ehrlacher and J. Huennekens, "Noble-gas broadening rates for the $6s^2\ {}^1S_0 \rightarrow 6s6p\ {}^{1,3}P_1$ resonance and intercombination lines of barium", Phys. Rev. A 47, 3097-3104 (1993).

E. Ehrlacher and J. Huennekens, "Excitation transfer among, and quenching of, the barium $6s5d\ {}^3D_J$ metastable levels due to collisions with argon, nitrogen, and barium perturbers", submitted to Phys. Rev. A.

E. Ehrlacher, R. Brown, and J. Huennekens, "Diffusion of $6s^2\ {}^1S_0$ ground state and 6s5d ${}^3D_J$ metastable level barium atoms through argon", manuscript in preparation.

Ph.D. Dissertations resulting from ARO grant DAAH04-93-G-0063

Edward A. Ehrlacher, "Collisional processes involving the metastable 6s5d ^3D_J levels of barium", Lehigh University, Dept. of Physics, 1993.

List of Participating Scientific Personnel - ARO grant DAAH04-93-G-0063

Principal Investigator

John Huennekens

Graduate Research Assistants

Raychel Brown
Ed Ehrlacher - Ph.D. August 1993
Cammie Gee
Jon Sagle

Undergraduate Research Participants

Amy Perkins